Structural and Electrochemical Studies of Li-ion Battery Cathode Single Particle by Using an *In-situ* FIB-SEM Setup and TXM Tomography

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Evolution of microstructures, the appearance of cracks and porosities, the formation of SEI at the electrode/electrolyte interface, and the transformation of crystal phases have to be properly investigated in order to get a better insight into the influence of charge/discharge processes in materials and the reaction mechanisms implied in electrochemical storage. In order to monitor microstructural evolution dynamically during electrochemical cycling, we developed a micro-scale battery set-up implemented within a FIB/SEM instrument. The single particle of cathode oxide (NCA and NMC) materials with a size of 5-10 µm is attached to a micromanipulator allowing to move the particle in the chamber and immersing in ionic liquid electrolyte (low vapor pressure), which is deposited on lithium metal. We studied structural modifications of individual particle after each charge/discharge cycle by FIB slicing and SEM imaging. We quantified the formation of cracks as a function of cycle number and extracted 2D skeleton and tortuosity. Evolution of the discharge capacity was correlated with cracks and porosities appearance inside cathode materials. Impedance measurements suggested an increase of lithium diffusion inside the particle that is relied on the crack formation. The changes of structural parameters such as porosity, grain connectivity and crack propagation that are induced by cycling were extracted from 3D reconstruction of both FIB tomography and TXM tomography and linked with electrochemical properties.